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**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371**

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U.S. APPLICATION NO (If known, see 37 C.F.R. 1.5)

09/701,142

INTERNATIONAL APPLICATION NO.
PCT/FR99/01210INTERNATIONAL FILING DATE
21 May 1999PRIORITY DATE CLAIMED
26 May 1999

TITLE OF INVENTION

ZINC ALUMINATE WITH HIGH SPECIFIC SURFACE AREA, PREPARATION METHOD AND USE FOR TREATING MOTOR VEHICLE EXHAUST GASES

APPLICANT(S) FOR DO/EO/US

Catherine HEDOUIN; Thierry SEGUELONG

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and the PCT Articles 22 and 39(1).
4. ☐ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☐ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☐ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern other document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☐ A FIRST preliminary amendment.
☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information: **PETITION FOR EXTENSION OF TIME**

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of)
)
Catherine HEDOUIN et al.) Group Art Unit: Unassigned
)
Application No.: Unassigned) Examiner: Unassigned
(Corresponds to PCT/FR99/01210))
)
International Filing Date: May 21, 1999)
)
For: ZINC ALUMINATE WITH HIGH)
SPECIFIC SURFACE AREA,)
PREPARATION METHOD AND USE)
FOR TREATING MOTOR VEHICLE)
EXHAUST GASES)

PRELIMINARY AMENDMENT

BOX PCT

Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

Prior to examination, please amend the above-captioned application as follows:

IN THE ABSTRACT:

Please delete the original abstract of the disclosure and insert the new abstract of the disclosure presented herewith on a separate sheet.

IN THE CLAIMS:

Kindly amend the claims as follows:

Claim 1, lines 1-2, change "characterized in that it has," to --having,--;
line 2, after "at" insert --least at--.

Claim 2, line 2, change "characterized in that it has," to --having,--.

Claim 3, lines 1-2, change "either of the preceding claims" to --claim 1--;
line 2, change "characterized in that it has," to --having--;
lines 4-5, delete ", more particularly at least 80 m²/g".

Claim 4, lines 1-2, change "one of the preceding claims" to --claim 1--;
line 2, change "characterized in that it has," to --having--;
lines 4-5, delete ", more particularly at least 70 m²/g".

Claim 5, lines 1-2, change "one of the preceding claims" to --claim 1--;
line 2, change "characterized in that it has," to --having--.

6. (Amended) Zinc aluminate according to [one of the preceding claims] claim 1, [characterized in that it comprises] comprising at least one additive [chosen] selected from the group consisting of the elements of Groups IA, IIA, VIIA to IB of the Periodic Table and from tin, gallium and the rare earths.

7. (Amended) Precursor composition for a zinc aluminate, [characterized in that it comprises] comprising compounds of zinc and [of] aluminium [and in that it is] which are capable of forming, after being calcined, a zinc aluminate, [this] said aluminate having, after being calcined at least at 800°C for 8 hours, a specific surface area of at least 85 m²/g.

Claim 8, line 2, change "characterized in that it" to --which--;
lines 4-5, delete ", more particularly at least 100 m²/g".

Claim 9, line 1, delete "or 8";
line 2, change "characterized in that it furthermore comprises" to
--comprising--;
line 3, change "chosen" to --selected-- and change "those" to --the
group consisting--.

10. (Amended) Process for the preparation of an aluminate according to [one of
claims 1 to 6, or of a precursor composition according to one of claims 7 to 9] claim 1,
[characterized in that it comprises] comprising the following steps:

- bringing a zinc salt, sol or alkoxide and an aluminium alkoxide [are brought]
together in a solvent medium, optionally with a salt, sol or alkoxide of at least one
[aforementioned] additive;
- hydrolysing the mixture thus formed [is hydrolysed] by adding water in an excess
amount with respect to the aluminium alkoxide;
- recovering the precipitate formed [is recovered] and optionally [dried] drying,
thereby obtaining the precursor composition;
- if required, [the] calcining said precipitate [is calcined], thereby obtaining the
aluminate.

11. (Amended) Process for the preparation of an aluminate comprising an additive according to claim 6 [or of a precursor composition comprising a compound of an element according to claim 9], [characterized in that it comprises] comprising the following steps:

- bringing a zinc salt, sol or alkoxide and an aluminium alkoxide [are brought] together in a solvent medium;
- hydrolyzing the mixture thus formed [is hydrolysed] by adding water in an excess amount with respect to the aluminium alkoxide;
- recovering the precipitate formed [is recovered] and optionally [dried] drying, thereby obtaining the precursor composition;
- if required, [the] calcining said precipitate [is calcined], thereby obtaining the aluminate;
- the precursor composition or the aluminate is impregnated with a solution of a salt of the additive or [of] the [aforementioned] element.

Claim 12, line 1, delete "or 11";

line 2, change "characterized in that" to --wherein--.

Claim 13, lines 1-2, change "11 or 12, characterized in that" to --wherein--.

Claim 14, lines 1-2, change "one of claims 10 to 13" to --claim 10--;

line 2, change "characterized in that" to --wherein--.

Claim 15, lines 1-2, change "one of claims 10 to 14" to --claim 10--;

line 2, change "characterized in that" to --wherein--.

Claim 16, lines 1-2, delete "the purpose of";

line 3, change "characterized in that" to --comprising using--;

line 4, change "one of claims 1 to 6 is used" to --claim 1--.

Claim 17, line 2, change "characterized in that" to --comprising using--;

lines 3-4, change "one of claims 1 to 6 is used" to --claim 1--.

18. (Amended) Process for the treatment of gases having a high oxygen content from motor vehicles, [characterized in that] comprising using a catalytic system comprising an aluminate according to [one of claims 1 to 6 is used, the gasses having a high oxygen content] claim 1.

19. (Amended) Catalytic system for the [implementation of a process according to claims 16, 17 or 18, characterized in that it comprises] treatment of gases for reducing the emissions of nitrogen oxides comprising an aluminate according to [one of claims 1 to 6] claim 1 on a substrate.

20. (Amended) [Use of an aluminate according to one of claims 1 to 6 or of a precursor composition according to one of claims 7 to 9 for the manufacture of a] A

catalytic system [according to claim 19] for the treatment of gases for reducing the emissions of nitrogen oxides comprising a precursor composition according to claim 7.

REMARKS

Entry of the foregoing amendment(s) is respectfully requested.

The claims have been amended to eliminate multiple dependency and to place them in better condition for U.S. patent practice.

Should the Examiner have any questions concerning the subject application, a telephone call to the undersigned would be appreciated.

Respectfully submitted,

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ZINC ALUMINATE HAVING A HIGH SPECIFIC SURFACE AREA, ITS
PROCESS OF PREPARATION AND ITS USE IN THE TREATMENT OF
EXHAUST GASES OF MOTOR VEHICLES

RHODIA CHIMIE

The present invention relates to a zinc aluminate which is characterized in that it has, after being calcined at 800°C for 8 hours, a specific surface area of at least 85 m²/g. The invention also relates to a precursor composition for this aluminate. The process for the preparation of the aluminate and of the composition comprises the following steps: a zinc salt, sol or alkoxide and an aluminium alkoxide are brought together in a solvent medium; the mixture thus formed is hydrolysed by adding water in an excess amount with respect to the aluminium alkoxide; the precipitate formed is recovered and optionally dried, thereby obtaining the precursor composition; if required, the said precipitate is calcined, thereby obtaining the aluminate. Finally, the invention relates to the use of the aluminate in a process for treating exhaust gases of motor vehicles.

ZINC ALUMINATE HAVING A HIGH SPECIFIC SURFACE AREA, ITS
PROCESS OF PREPARATION AND ITS USE IN THE TREATMENT OF
EXHAUST GASES OF MOTOR VEHICLES

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RHODIA CHIMIE

The present invention relates to a zinc
10 aluminate having a high specific surface area, to a
precursor composition for this aluminate, to a process
for the preparation of this aluminate and of this
composition, and to the use of the aluminate in a process
for treating gases, especially exhaust gases of motor
15 vehicles.

It is known that the reduction of emissions of
nitrogen oxides (NO_x) from the exhaust gases of
motor-vehicle engines or of industrial plants constitutes
a major environmental protection problem. In the case of
20 motor vehicles, "three-way" catalysts are particularly
employed, which catalysts use stoichiometrically the
reducing gases present in the mixture. However, any
excess oxygen results in a sudden deterioration in the
performance of the catalyst.

25 However, some engines, such as diesel engines
or petrol engines operating in lean burn mode, are
sparing in terms of fuel but emit exhaust gases which
permanently contain a large excess of oxygen, for example

at least 5%. A standard three-way catalyst is therefore of no effect on the NO_x emissions from these engines. Moreover, the tightening of the standards with regard to motor vehicle postcombustion, which now extend to these
5 types of engines, makes it absolutely essential to limit NO_x emissions.

For these engines, catalysts in spinel form, based on aluminium and zinc, have been proposed. However, these catalysts must be further improved since they do
10 not have very high specific surface areas at high temperature. Now, surface stability, i.e. the fact of being able to maintain a high surface area at high temperature, is one aspect likely to improve the performance of catalysts.

15 There is therefore a need for a spinel-type catalyst having a high specific surface area.

To this end, the zinc aluminate of the invention is characterized in that it has, after being calcined at 800°C for 8 hours, a specific surface area
20 of at least 85 m²/g.

The invention also relates to a precursor composition for a zinc aluminate, which composition is characterized in that it comprises compounds of zinc and of aluminium and in that it is capable of forming, after
25 being calcined, a zinc aluminate, this aluminate having, after being calcined at 800°C for 8 hours, a specific surface area of at least 85 m²/g.

Another subject of the invention is a process

for the preparation of an aluminate or of a composition of the type described above, which process is characterized in that it comprises the following steps:

- a zinc salt and an aluminium alkoxide are brought together in a solvent medium;

- the mixture thus formed is hydrolysed by adding water in an excess amount with respect to the aluminium alkoxide;

- the precipitate formed is recovered and optionally dried, thereby obtaining the precursor composition;

- if required, the said precipitate is calcined, thereby obtaining the aluminate.

Further characteristics, details and advantages of the invention will become even more apparent on reading the description which follows, together with the various concrete but non-limiting examples intended to illustrate it.

The aluminate of the invention is a zinc aluminate. It has a structure of the spinel type, ZnAl_2O_4 . It may be in the form of one or more phases which are zinc-depleted or zinc-enriched with respect to ZnAl_2O_4 , these phases satisfying the formulae $\text{Zn}_{1-x}\text{Al}_2\text{O}_{4-\delta}$ and $\text{Zn}_{1+x}\text{Al}_2\text{O}_{4+\delta}$, where $0 < x \leq 0.95$. The values of x may more particularly satisfy the following relationships: $0 < x \leq 0.85$, $0 < x \leq 0.8$ and more particularly $0 < x \leq 0.5$. Finally, x may satisfy the relationship $0.4 \leq x \leq 0.85$. The aluminate may furthermore contain one

or more additives. These additives may be chosen from the elements of Groups IA, IIA, VIIA to IB of the Periodic Table and from tin, gallium and the rare earths.

The Periodic Table of the Elements to which
5 reference is made is the one published in the Supplément
au Bulletin de la Société Chimique de France (*Supplement
to the Bulletin of the Chemical Society of France*) No. 1
(January 1966). Moreover, the term "rare earths" should
be understood to mean the elements of the group formed by
10 yttrium and the elements of the Periodic Table having an
atomic number of between 57 and 71 inclusive.

As element of Group VIIA, mention may more
particularly be made of manganese; as element of Group
VIII, mention may especially be made of iron; as elements
15 of Group IB, mention may more particularly be made of
copper and silver.

These additives may especially be present in
the aluminate as partial substitution for the zinc or for
the aluminium.

20 One characteristic of the aluminate of the
invention is its specific surface area. The term
"specific surface area" should be understood to mean
here, and for the rest of the description, the BET
specific surface area determined by nitrogen adsorption
25 in accordance with the ASTM D 3663-78 standard
established from the Brunauer - Emmett - Teller method
described in the publication "The Journal of the American
Chemical Society, 60, 309 (1938)".

Even after being calcined at high temperature, the aluminate of the invention still has a high value of surface area. Thus, after calcining at 800°C for 8 hours, this specific surface area is at least 85 m²/g. It may be at least 90 m²/g and more particularly at least 100 m²/g, still after calcining at 800°C for 8 hours. Values of at least 120 m²/g may be achieved.

High values of this surface area are maintained at even higher temperatures since the aluminate of the invention may have, after being calcined at 900°C for 2 hours, a specific surface area of at least 70 m²/g, more particularly at least 80 m²/g. Furthermore, at 1000°C after calcining for 6 hours, specific surface areas of at least 50 m²/g, more particularly at least 70 m²/g may be observed. This means that the surface area of the aluminate is stable over a wide temperature range. The values which have just been given above extend to calcining in air. In addition, it may be noted that the aluminate of the invention has a high ageing resistance. What is meant by this is that, under particular calcining conditions, its specific surface area varies relatively little. Thus, after calcining for 6 hours at 1000°C in an H₂O/N₂ medium with 10% H₂O by volume, the surface area remains substantially the same as that obtained after calcining in air at the same temperature and for the same duration, namely at least 50 m²/g. The same results are obtained for calcining in an O₂/H₂O/N₂ medium with 10% H₂O and 10% O₂ by volume.

Moreover, the aluminate of the invention may have a pore volume of at least 0.6 ml/g, this porosity being determined by mercury intrusion porosimetry. The measurements were made using a Micromeretics Auto Pore 5 9220 machine on powders degassed overnight in an oven heated at 200°C. The operating parameters are as follows: penetrometer constant : 21.63, capillary volume : 1.1, contact angle : 140°. The porosity may more particularly be at least 2 ml/g and may be, for example, between 2.5 10 and 3.5 ml/g.

The invention also relates to a precursor composition for the aluminate, such as the one that has just been described above.

This composition comprises compounds of zinc 15 and of aluminium and, if required, compounds of the abovementioned additives. The main characteristic of the precursor composition is its ability, after being calcined, to give a zinc aluminate. The calcining temperature above which the aluminate forms is 20 approximately 500°C. Furthermore, the aluminate thus obtained has the characteristics given above, that is to say that, if it is calcined at a temperature of 800°C for 8 hours, it retains a high specific surface area of at least 85 m²/g, more particularly at least 90 m²/g and even 25 more particularly at least 100 m²/g. Of course, all the surface area values given above with regard to the aluminate at temperatures of 800°C and 900°C also apply here.

The process for the preparation of the aluminate and of its precursor composition will now be described.

The first step of this process consists in
5 bringing a zinc salt, sol or alkoxide and an aluminium
alkoxide together in a solvent medium, optionally with
a salt, sol or alkoxide of at least one aforementioned
additive. The zinc salt or alkoxide, as well as the salt
or alkoxide of the additive, must be soluble in the
10 solvent medium. The zinc salt or the salt of the additive
is, for example, an inorganic salt such as a nitrate or
a chloride or else an organic salt such as a citrate, an
oxalate or an acetate. The aluminium alkoxide may, for
example, be an ethoxide, a butoxide or an isopropoxide.

15 The solvent medium is chosen from any medium
in which the zinc salt or alkoxide and the aluminium
alkoxide are soluble. Generally, an alcoholic solvent is
used. As alcoholic solvent, mention may be made of
saturated monoalcohols and more particularly those having
20 a short chain (for example at most C_8), such as methanol,
ethanol, propanol and butanol. It is also possible to use
unsaturated alcohols and polyalcohols, such as, for
example, ethylene glycol, propylene glycol, hexylene
glycol, propanediol and butanediol. A ketone such as
25 acetylacetone may also be used.

The reactants may be brought together in the
solvent medium in any manner. However, according to one
particular embodiment of the invention, the zinc salt and

the aluminium alkoxide are brought together by adding to the aluminium alkoxide, which constitutes a stock, the zinc salt in the solvent medium, i.e. dissolved beforehand in this medium.

5 It will be noted here, according to another version of the invention, that it is possible to heat the mixture thus obtained. This makes it easier for the salts to dissolve and allows the next step - hydrolysis and precipitation - to be better controlled.

10 The second step of the process of the invention consists in hydrolysing the mixture obtained from the previous step.

 This hydrolysis takes place by adding water to the mixture. According to one characteristic of the
15 process of the invention, the hydrolysis is carried out by using an excess amount of water with respect to the aluminium alkoxide. This excess is determined by the H_2O /aluminium alkoxide molar ratio. In general, this ratio may be at least 6, more particularly at least 10 and even
20 more particularly at least 20. However, in the case of the preparation of a zinc-depleted aluminate, this ratio may be lower. More specifically, in the case of the preparation of an aluminate having a Zn/Al ratio of less than 0.4, the H_2O /aluminium alkoxide molar ratio may be at
25 least 3, more particularly at least 4.

 The water may be provided in the form of a water-alcohol mixture, it being possible for the alcohol to be especially chosen from those mentioned above with

regard to the solvent medium. Mention may more particularly be made of ethanol.

The hydrolysis results in the formation of a precipitate of the elements.

5 The precipitate obtained is separated from the reaction mixture by any known means, especially by centrifuging.

If necessary, the precipitate may be washed.

Optionally, the precipitate may then be dried.

10 At this stage, the precursor composition of the invention is obtained. The aluminate is prepared by calcining the precipitate (precursor composition) at a temperature of at least 500°C.

 Mention may be made of another process for the
15 preparation of an aluminate or of a precursor according to the invention if this aluminate or precursor comprises an additive of the type mentioned above. This process consists in adding the additive not during the synthesis of the aluminate but by impregnating either the precursor
20 composition, for example the dried precipitate, or the aluminate itself, i.e. the calcined precipitate. The impregnation is carried out using a solution of a salt of the additive of the type given above for example.

 Dry impregnation is more particularly used. Dry
25 impregnation consists in adding to the product to be impregnated a volume of an aqueous solution of the element which is equal to the pore volume of the solid to be impregnated.

Moreover, the invention relates to a process for the treatment of exhaust gases of motor vehicles, in which a catalytic system comprising an aluminate as described above is used.

5 The invention also relates to a process in which a catalytic system comprising this same aluminate is used for the treatment of gases which may comprise nitrogen oxides for the purpose of reducing the emissions of these nitrogen oxides.

10 In this case, the gases capable of being treated are, for example, those coming from gas turbines, from boilers of thermal power stations or else from internal combustion engines. In the latter case, these may be especially diesel engines or engines operating in
15 lean burn mode.

 The aluminate of the invention thus applies to the treatment of gases having a high oxygen content and containing nitrogen oxides, for the purpose of reducing the emissions of these oxides. The expression "gases
20 having a high oxygen content" should be understood to mean gases having an excess of oxygen with respect to the amount necessary for stoichiometric combustion of the fuels and, more particularly, gases permanently having an excess of oxygen with respect to the stoichiometric value
25 $\lambda = 1$. The value λ is correlated to the air/fuel ratio in a manner known per se, especially a manner known in the field of internal combustion engines. In other words, the aluminate of the invention applies to the treatment of

gases coming from systems of the type described in the previous paragraph and operating permanently under conditions such that λ is always strictly greater than 1. In the case of gases having a high oxygen content, the aluminate of the invention thus applies, on the one hand, to the treatment of the gases from an engine which is operating in lean burn mode and which has a high oxygen content (expressed by volume) of generally between 2.5 and 5% and, on the other hand, to the treatment of gases which have an even higher oxygen content, for example the gases from diesel-type engines, i.e. at least 5% or more than 5%, more particularly at least 10%, it being possible, for example, for this content to be between 5 and 20%.

The gases contain a reducing agent which may be one or more hydrocarbons and one of the reactions that it is desired to catalyse in this case is the HC (hydrocarbons) + NO_x reaction.

The hydrocarbons which may be used as reducing agent for eliminating the NO_x are especially gases or liquids from the families of saturated carbides, ethylenic carbides, acetylenic carbides, aromatic carbides and hydrocarbons of petroleum cuts such as, for example, methane, ethane, propane, butane, pentane, hexane, ethylene, propylene, acetylene, butadiene, benzene, toluene, xylene, kerosene and gas oil.

The gases may also contain, as reducing agent, organic compounds containing oxygen. These compounds may

especially be alcohols, for example of the saturated alcohol type, such as methanol, ethanol or propanol; ethers such as methyl ether or ethyl ether; esters such as methyl acetate and ketones.

5 The gases may also contain ammonia as a reducing agent.

 In this application to the treatment of gases, the aluminate may be used in catalytic compositions which may be in various forms, such as granules, beads, cylinders or a honeycomb of varying dimensions, it being
10 possible for these compositions to comprise the aluminate of the invention on any support normally used in the field of catalysis, such as, for example, ZrO_2 , Al_2O_3 , TiO_2 , CeO_2 , SiO_2 or mixtures thereof.

15 The invention also relates more particularly to a catalytic system for the gas treatment processes described above. This system is characterized in that it comprises an aluminate on a substrate. Such a system generally comprises a wash coat incorporating the
20 aluminate and a support of the type described above, the wash coat being deposited on a substrate, for example a substrate of the metal or ceramic monolith type.

 The systems are mounted in a known manner in the exhaust pipes of vehicles in the case of the
25 application to the treatment of exhaust gases.

 Finally, the invention also relates to the use of an aluminate or of a precursor composition, such as those described above, for the manufacture of such a

catalytic system. Examples will now be given.

EXAMPLE 1

This example relates to the preparation of
5 ZnAl_2O_4 .

The following raw materials are used:

- Crystallized 99% pure $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, molecular
weight: 297.47

- 97% pure aluminium tri-*sec*-butoxide
10 $(\text{C}_2\text{H}_5\text{CH}_2\text{CH}_2\text{O})_3\text{Al}$

- 99% pure hexylene glycol (2-methyl
2,4-pentanediol)

- absolute ethanol, molecular weight:
46.07; $d: 0.79 \text{ g/cm}^3$.

15 1.25 mol of the zinc salt are dissolved in 1 l
of hexylene glycol. This solution is added to 2.5 mol of
aluminium tri-*sec*-butoxide introduced beforehand into the
reactor, in a single go and with vigorous stirring
(500 rpm). The mixture is heated up to 70°C and kept at
20 this temperature for 2 hours. Next, a water/ethanol
(50/50 by volume) mixture is added at a rate of
5 ml/minute. The water/aluminium tri-*sec*-butoxide ratio
is equal to 28. The mixture is left to cool overnight
with stirring. The precipitate obtained is separated by
25 centrifuging. The precipitate is oven-dried as a thin
layer at 70°C for 48 h. Finally, the product is calcined.
The temperature rise is 5°C/minute . Next, the temperature
is maintained at a constant level with the value given

above and for the times given above.

After calcining for 6 hours at 600°C, the specific surface area of the product is 136 m²/g.

After calcining for 6 hours at 700°C, the
5 specific surface area of the product is 125 m²/g.

After calcining for 8 hours at 800°C, the specific surface area of the product is 115 m²/g.

After calcining for 2 hours at 900°C, the specific surface area of the product is 101 m²/g.

10 After calcining for 2 hours at 1000°C, the specific surface area of the product is 76 m²/g and it is 53 m²/g after 6 hours at the same temperature.

After calcining for 6 hours at 1000°C in an H₂O/N₂ mixture with 10% H₂O by volume, the surface area
15 remains equal to 53 m²/g. The same result is obtained for calcining for 6 hours at 1000°C in an O₂/H₂O/N₂ medium with 10% H₂O and 10% O₂ by volume.

EXAMPLE 2

20 The precipitate obtained in Example 1 is dried for 48 h at 70°C. Next, it is impregnated with a solution of SnCl₂·2H₂O dissolved in ethanol. The technique used is dry impregnation. The amount of Sn deposited is equal to 1.6% by weight with respect to the zinc aluminate oxide.
25 The product thus obtained is oven-dried for 2 h at 110°C and then calcined for 8 h at 800°C (rate of rise, 5°C/minute).

The specific surface area of the product thus

obtained is equal to 115 m²/g.

EXAMPLE 3

This example relates to the preparation of
5 ZnAl_{1.8}Ga_{0.2}O₄.

The same raw materials as in Example 1 are used with, in addition, a 1.807 mol/l solution of Ga(NO₃)₃, d = 1.365 g/cm³.

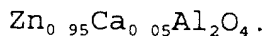
1.25 mol of the zinc salt are dissolved in
10 1 litre of hexylene glycol into which 0.25 mol of gallium nitrate is then introduced.

This solution is added to 2.25 mol of aluminium tri-sec-butoxide introduced beforehand into the reactor, with vigorous stirring (500 rpm). The mixture is heated
15 to 70°C and maintained at this temperature for 2 hours. A water/ethanol (50/50 by volume) mixture is then added at a rate of 5 ml/minute. The water/aluminium tri-sec-butoxide ratio is equal to 25. The mixture is left to cool overnight with stirring. The precipitate
20 obtained is separated by centrifuging. The precipitate obtained is dried as a thin layer at 70°C for 48 h and then calcined for 8 hours at 800°C (rate of rise 5°C/minute).

The specific surface area of the product thus
25 obtained is equal to 113 m²/g.

EXAMPLE 4

This example relates to the preparation of



The same raw materials as in Example 1 are used with, in addition, a solution of 98% pure $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$.

1.19 mol of the zinc salt are dissolved in 1 litre of hexylene glycol into which 0.06 mol of calcium nitrate is then introduced.

This solution is added to 2.5 mol of aluminium tri-sec-butoxide introduced beforehand into the reactor, with vigorous stirring (500 rpm). The mixture is heated to 70°C and maintained at this temperature for 2 hours. A water/ethanol (50/50 by volume) mixture is then added at a rate of 5 ml/minute. The water/aluminium tri-sec-butoxide ratio is equal to 28. The mixture is left to cool overnight with stirring. The precipitate obtained is separated by centrifuging. The precipitate obtained is dried as a thin layer at 70°C for 48 h and then calcined for 8 hours at 800°C (rate of rise 5°C/minute).

The specific surface area of the product thus obtained is equal to 119 m²/g.

EXAMPLE 5

This example relates to the preparation of $\text{Zn}_{0.95}\text{Li}_{0.05}\text{Al}_2\text{O}_4$.

The same raw materials as in Example 1 are used with, in addition, a solution of 99% pure $\text{Li}(\text{NO}_3)$.

1.19 mol of the zinc salt are dissolved in 1 litre of hexylene glycol into which 0.06 mol of lithium

nitrate is then introduced.

This solution is added to 2.5 mol of aluminium tri-sec-butoxide introduced beforehand into the reactor, with vigorous stirring (500 rpm). The mixture is heated
5 to 70°C and maintained at this temperature for 2 hours. A water/ethanol (50/50 by volume) mixture is then added at a rate of 5 ml/minute. The water/aluminium tri-sec-butoxide ratio is equal to 28. The mixture is left to cool overnight with stirring. The precipitate
10 obtained is separated by centrifuging. The precipitate obtained is dried as a thin layer at 70°C for 48 h and then calcined for 8 hours at 800°C (rate of rise 5°C/minute).

The specific surface area of the product thus
15 obtained is equal to 108 m²/g.

EXAMPLE 6

This example relates to an aluminate of formula ZnAl₂O₄ which comprises silver as additive.

20 The precipitate obtained in Example 1 is dried for 48 h at 70°C. Next, it is impregnated with a solution of AgNO₃ (99.8%). The technique used is dry impregnation. The amount of silver deposited is equal to 1.6% by weight with respect to the zinc aluminate oxide. The product
25 thus obtained is oven dried for 2 h at 110°C and then calcined for 8 h at 800°C (rate of rise 5°C/minute).

The specific surface area of the product thus obtained is equal to 90 m²/g.

EXAMPLE 7

The process as in Example 1 is carried out in order to prepare zinc aluminates having different Zn/Al ratios. The same raw materials are used, in the necessary proportions. For the products 7-1 to 7-3, the H₂O/aluminium alkoxide molar ratio is 28. For the products 7-4 to 7-6, this ratio is 4. The characteristics of the products prepared are given below.

Products	Formulae	Specific surface area (8 h at 800°C) in m ² /g
7-1	Zn _{1.05} Al ₂ O ₄	88
7-2	Zn _{0.95} Al ₂ O ₄	92
7-3	Zn _{0.8} Al ₂ O ₄	140
7-4	Zn _{0.6} Al ₂ O ₄	102
7-5	Zn _{0.4} Al ₂ O ₄	139
7-6	Zn _{0.3} Al ₂ O ₄	110

EXAMPLE 8

In this example, the products obtained in the previous examples are tested in order to evaluate their catalytic performance.

A quartz reactor is charged with 0.2 g of the catalyst powder. The powder used was precompacted and then ground and screened so as to isolate the particle size range between 0.125 and 0.250 mm.

The reaction mixture entering the reactor has the following composition (by volume):

- NO = 300 ppmv
- C₃H₆ = 150 ppmv or 450 ppmv
- C₃H₈ = 150 ppmv or 450 ppmv
- CO = 350 ppmv
- 5 - O₂ = 10%
- CO₂ = 10%
- H₂O = 10%
- N₂ = qsp 100%

The overall flow rate is 30 Nl/h.

10 The HSV is about 200,000 h⁻¹.

The HC(C₃H₆ + C₃H₈), NO and NO_x (NO_x = NO + NO₂) signals are continuously recorded, as is the temperature in the reactor.

15 The HC signal is given by a total HC Beckman detector based on the principle of detection using flame ionization.

20 The NO and NO_x signals are given by an Ecophysics NO_x analyser based on the principle of chemiluminescence: it gives the NO, NO_x and NO₂ values, the latter being calculated from the difference between the NO_x and NO signals.

25 The catalytic activity is measured from the HC, NO and NO_x signals as a function of temperature during a programmed temperature rise of 150 to 700°C at a rate of 15°C/minute and from the following equations:

- The degree of conversion of NO (TNO) in %, which is given by:

$$TNO = 100 (NO^0 - NO) / NO^0$$

where NO^0 is the NO signal at the time $t = 0$, which corresponds to the NO signal obtained with the reaction mixture by-passing the catalytic reactor and NO is the NO signal at the time t .

- 5 - The degree of conversion of HC (THC) in %, which is given by:

$$\text{THC} = 100 (\text{HC}^0 - \text{HC}) / \text{HC}^0$$

where HC^0 is the HC signal at the time $t = 0$, which corresponds to the HC signal obtained with the reaction mixture by-passing the catalytic reactor and HC is the
10 HC signal at the time t .

- The degree of conversion of NO_x (TNO_x) in %, which is given by:

$$\text{TNO}_x = 100 (\text{NO}_x^0 - \text{NO}_x) / \text{NO}_x^0$$

- 15 where NO_x^0 is the NO_x signal at the time $t = 0$, which corresponds to the NO_x signal obtained with the reaction mixture by-passing the catalytic reactor and NO_x is the NO_x signal at the time t .

Table 1 below gives the results obtained with the product of Example 1 calcined for 8 h at 800°C and for a reaction mixture in which NO = 300 ppmv, C₃H₆ = C₃H₈ = 150 ppmv, i.e. an HC₁/NO ratio of 3 (HC₁ being expressed as the carbon number, i.e. here 6 x 150/300).

Table 1

Temperature (°C)	THC (%)	TNO (%)	TNO _x (%)
350	0.6	1.1	1.3
400	1	2.1	2
450	6	8.8	7.9
500	24.8	23.3	23.7
550	46.4	44.5	44.3
600	75.6	59.5	59.4
650	99	28	25.8

Table 2 below gives the results obtained with the product of Example 1 calcined for 8 h at 800°C, for a reaction mixture in which NO = 300 ppmv, C₃H₆ = C₃H₈ = 450 ppmv, i.e. an HC₁/NO ratio of 9.

Table 2

10	Temperature (°C)	THC (%)	TNO (%)	TNO_x (%)^x
	350	0.8	0.2	1.4
	400	1.8	0	1.4
	450	7.5	4.8	6.2
15	500	29	32.1	32.8
	550	50.9	71.5	70.9
	600	71.9	87.9	86.4
	650	95	69	63.9

Table 3 below gives the results obtained with the product of Example 1 calcined for 2 h at 900°C and for the same reaction mixture as in Example 1, i.e. an HC₁/NO ratio of 3.

Table 3

10	Temperature (°C)	THC (%)	TNO (%)	TNO_x (%)
	350	0.3	0	0
	400	0.7	0	0
	450	2.1	1.2	2
15	500	10.5	11	11.3
	550	3.1	28.2	28.5
	600	68.6	52.6	52.9
	650	97.9	28.7	27.5

Table 4 below gives the results obtained with the product of Example 1 calcined for 2 h at 1000°C and for the same reaction mixture as in Example 1, i.e. an HC₁/NO ratio of 3.

Table 4

10	Temperature (°C)	THC (%)	TNO (%)	TNO_x (%)
	350	0	0	0
	400	0	0	0
	450	1.3	1.5	1.2
15	500	9.4	12.5	12.2
	550	34.1	37.6	37.5
	600	80.9	51.7	51.2
	650	100	21.5	18.3

Table 5 below gives the results obtained with the product of Example 2 calcined for 2 h at 800°C and for the same reaction mixture as in the first case of the product of Example 1, i.e. an HC₁/NO ratio of 3.

Table 5

10	Temperature (°C)	THC (%)	TNO (%)	TNO_x (%)
	350	0	2.2	2.6
	400	0.9	3.3	3.7
	450	5.6	10.9	11.2
15	500	25	32	32.2
	550	49	38.4	38.1
	600	63.3	31.1	28.6
	650	87.4	26.8	21.2

Table 6 below gives the results obtained with the product of Example 2 calcined for 2 h at 800°C for a reaction mixture in which NO = 300 ppmv, C₃H₆ = C₃H₈ = 450 ppmv, i.e. an HC_i/NO ratio of 9.

Table 6

10	Temperature (°C)	THC (%)	TNO (%)	TNO _x (%)
	350	2.6	1.3	1.3
	400	6.1	3.8	3.2
	450	13.9	12.5	12.4
15	500	35.1	47.3	47.3
	550	55.4	54.4	54.1
	600	71.8	42	41.7
	650	96.7	31.9	29.8

Table 7 below gives the results obtained with the product of Example 7 calcined for 2 h at 800°C and for a reaction mixture with an HC_1/NO ratio of 3. Furthermore, the product is used in an amount of 100 mg of powder with 100 mg of SiC.

Table 7

10	Temperature (°C)	THC (%)	TNO (%)	TNO_x (%)^x
	350	2.6	2.1	2.6
	400	2.9	1.4	2.1
15	450	4.8	4.2	4.2
	500	21.1	26.6	26.2
	550	63	46.4	45
	600	90	34.5	30.1
20	650	98.9	24.5	7

Table 8 below gives the maximum % NO_x values obtained, and the corresponding temperature, with products of Example 8 that are calcined for 2 h at 800°C and for a reaction mixture having an HC₁/NO ratio of 3. Furthermore, the products are used in an amount of 100 mg of powder with 100 mg of SiC.

Table 8

Products	% NO _x and temperature
7-3	35% 620°C
7-4	38% 625°C
7-5	42% 600°C
7-6	46% 600°C

CLAIMS

1. Zinc aluminate, characterized in that it has, after being calcined at 800°C for 8 hours, a
5 specific surface area of at least 85 m²/g.

2. Zinc aluminate according to claim 1, characterized in that it has, after being calcined at 800°C for 8 hours, a specific surface area of at least 100 m²/g.

10 3. Zinc aluminate according to either of the preceding claims, characterized in that it has, after being calcined at 900°C for 2 hours, a specific surface area of at least 70 m²/g, more particularly at least 80 m²/g.

15 4. Zinc aluminate according to one of the preceding claims, characterized in that it has, after being calcined at 1000°C for 6 hours, a specific surface area of at least 50 m²/g, more particularly at least 70 m²/g.

20 5. Zinc aluminate according to one of the preceding claims, characterized in that it has, after being calcined at 1000°C for 6 hours, in an H₂O/N₂ medium with 10% H₂O by volume, a specific surface area of at least 50 m²/g.

25 6. Zinc aluminate according to one of the preceding claims, characterized in that it comprises at least one additive chosen from the elements of Groups IA, IIA, VIIA to IB of the Periodic Table and from tin,

gallium and the rare earths.

7. Precursor composition for a zinc aluminate, characterized in that it comprises compounds of zinc and of aluminium and in that it is capable of forming, after being calcined, a zinc aluminate, this aluminate having, after being calcined at 800°C for 8 hours, a specific surface area of at least 85 m²/g.

8. Composition according to claim 7, characterized in that it is capable of forming an aluminate having, after being calcined at 800°C for 8 hours, a specific surface area of at least 90 m²/g, more particularly at least 100 m²/g.

9. Composition according to claim 7 or 8, characterized in that it furthermore comprises at least one compound of an element chosen from those of Groups IA, IIA, VIIA to IB of the Periodic Table and from tin, gallium and the rare earths.

10. Process for the preparation of an aluminate according to one of claims 1 to 6, or of a precursor composition according to one of claims 7 to 9, characterized in that it comprises the following steps:

- a zinc salt, sol or alkoxide and an aluminium alkoxide are brought together in a solvent medium, optionally with a salt, sol or alkoxide of at least one aforementioned additive;

- the mixture thus formed is hydrolysed by adding water in an excess amount with respect to the aluminium alkoxide;

- the precipitate formed is recovered and optionally dried, thereby obtaining the precursor composition;

- if required, the said precipitate is calcined,
5 thereby obtaining the aluminate.

11. Process for the preparation of an aluminate comprising an additive according to claim 6 or of a precursor composition comprising a compound of an element according to claim 9, characterized in that it
10 comprises the following steps:

- a zinc salt, sol or alkoxide and an aluminium alkoxide are brought together in a solvent medium;

- the mixture thus formed is hydrolysed by adding water in an excess amount with respect to the aluminium
15 alkoxide;

- the precipitate formed is recovered and optionally dried, thereby obtaining the precursor composition;

- if required, the said precipitate is calcined,
20 thereby obtaining the aluminate;

- the precursor composition or the aluminate is impregnated with a solution of a salt of the additive or of the aforementioned element.

12. Process according to claim 10 or 11,
25 characterized in that an alcoholic solvent is used as solvent medium.

13. Process according to claim 10, 11 or 12, characterized in that water in the form of a

water-alcohol mixture is added.

14. Process according to one of claims 10 to 13, characterized in that the zinc salt and the aluminium alkoxide are brought together by adding the zinc salt in the solvent medium to the aluminium alkoxide.

15. Process according to one of claims 10 to 14, characterized in that the precipitate is calcined at a temperature of at least 500°C.

16. Process for the treatment of gases for the purpose of reducing the emissions of nitrogen oxides, characterized in that a catalytic system comprising an aluminate according to one of claims 1 to 6 is used.

17. Process for the treatment of exhaust gases of motor vehicles, characterized in that a catalytic system comprising an aluminate according to one of claims 1 to 6 is used.

18. Process for the treatment of gases from motor vehicles, characterized in that a catalytic system comprising an aluminate according to one of claims 1 to 6 is used, the gases having a high oxygen content.

19. Catalytic system for the implementation of a process according to claims 16, 17 or 18, characterized in that it comprises an aluminate according to one of claims 1 to 6 on a substrate.

20. Use of an aluminate according to one of claims 1 to 6 or of a precursor composition according to one of claims 7 to 9 for the manufacture of a catalytic system according to claim 19.

Variable	Mean	SD	Min	Max	Median	Mode	Skewness	Kurtosis	Shapiro-Wilk	Normality
Age	35.2	12.5	18	65	32	30	0.15	2.10	0.98	Normal
Gender	1.2	0.4	1	2	1	1	0.05	0.10	0.99	Normal
Marital Status	2.1	0.8	1	3	2	2	0.10	0.50	0.97	Normal
Education	14.5	2.5	10	20	15	15	0.05	0.10	0.99	Normal
Income	1200	300	500	2500	1100	1000	0.10	0.50	0.97	Normal
Occupation	1.5	0.5	1	3	1	1	0.05	0.10	0.99	Normal
Health Status	2.5	0.5	1	3	2	2	0.05	0.10	0.99	Normal
Stress Level	3.5	1.0	1	5	3	3	0.10	0.50	0.97	Normal
Life Satisfaction	4.0	0.8	1	5	4	4	0.05	0.10	0.99	Normal
Work-Life Balance	3.0	0.7	1	4	3	3	0.05	0.10	0.99	Normal
Family Support	4.5	0.6	1	5	4	4	0.05	0.10	0.99	Normal
Community Involvement	2.0	0.5	1	3	2	2	0.05	0.10	0.99	Normal
Personal Growth	3.8	0.9	1	5	3	3	0.10	0.50	0.97	Normal
Overall Well-being	4.2	0.7	1	5	4	4	0.05	0.10	0.99	Normal

Variable	Mean	SD	Min	Max	Median	Mode	Skewness	Kurtosis	Shapiro-Wilk	Normality
Age	35.2	12.5	18	65	32	30	0.15	2.10	0.98	Normal
Gender	1.2	0.4	1	2	1	1	0.05	0.10	0.99	Normal
Marital Status	2.1	0.8	1	3	2	2	0.10	0.50	0.97	Normal
Education	14.5	2.5	10	20	15	15	0.05	0.10	0.99	Normal
Income	1200	300	500	2500	1100	1000	0.10	0.50	0.97	Normal
Occupation	1.5	0.5	1	3	1	1	0.05	0.10	0.99	Normal
Health Status	2.5	0.5	1	3	2	2	0.05	0.10	0.99	Normal
Stress Level	3.5	1.0	1	5	3	3	0.10	0.50	0.97	Normal
Life Satisfaction	4.0	0.8	1	5	4	4	0.05	0.10	0.99	Normal
Resilience	3.0	0.5	1	4	3	3	0.05	0.10	0.99	Normal
Optimism	3.5	0.5	1	4	3	3	0.05	0.10	0.99	Normal
Emotional Stability	3.0	0.5	1	4	3	3	0.05	0.10	0.99	Normal
Self-Esteem	3.5	0.5	1	4	3	3	0.05	0.10	0.99	Normal
Life Satisfaction	4.0	0.8	1	5	4	4	0.05	0.10	0.99	Normal
Resilience	3.0	0.5	1	4	3	3	0.05	0.10	0.99	Normal
Optimism	3.5	0.5	1	4	3	3	0.05	0.10	0.99	Normal
Emotional Stability	3.0	0.5	1	4	3	3	0.05	0.10	0.99	Normal
Self-Esteem	3.5	0.5	1	4	3	3	0.05	0.10	0.99	Normal

R98091

COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY (Includes Reference to Provisional and PCT International Applications)	Attorney's Docket No. 022701-892
--	-------------------------------------

As a below named inventor, I hereby declare that:
 My residence, post office address and citizenship are as stated below next to my name;
 I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

ZINC ALUMINATE WITH HIGH SPECIFIC SURFACE AREA, PREPARATION METHOD AND USE FOR
TREATING MOTOR VEHICLE EXHAUST GASES

the specification of which (check only one item below):

- ☐ is attached hereto.
- ☐ was filed as United States application
 Number _____
 on _____
 and was amended
 on _____ (if applicable).
- ☒ was filed as PCT international application
 Number PCT/FR99/01210
 on 21 May 1999
 and was amended
 on _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 (a)-(e) of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed:

PRIOR FOREIGN/PCT APPLICATION(S) AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. § 119:

COUNTRY (if PCT, indicate "PCT")	APPLICATION NUMBER	DATE OF FILING (day, month, year)	PRIORITY CLAIMED UNDER 35 U.S.C. § 119
FR	98/06610	26 May 1998	<u>X</u> Yes <u> </u> No
			<u> </u> Yes <u> </u> No
			<u> </u> Yes <u> </u> No
			<u> </u> Yes <u> </u> No
			<u> </u> Yes <u> </u> No

I hereby claim the benefit under Title 35, United States Code § 119(e) of any United States provisional application(s) listed below.

_____	_____
(Application Number)	(Filing Date)
_____	_____
(Application Number)	(Filing Date)

COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY (CONT'D)
(Includes Reference to Provisional and PCT International Applications)

Attorney's Docket No.

022701-892

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose to the Office all information known to me to be material to the patentability as defined in Title 37, Code of Federal Regulations §1.56, which became available between the filing date of the prior application(s) and the national or PCT international filing date of this application:

PRIOR U.S. APPLICATIONS OR PCT INTERNATIONAL APPLICATIONS DESIGNATING THE U.S. FOR BENEFIT UNDER 35 U.S.C. §120:

U.S. APPLICATIONS		STATUS (check one)		
U.S. APPLICATION NUMBER	U.S. FILING DATE	PATENTED	PENDING	ABANDONED
PCT APPLICATIONS DESIGNATING THE U.S.				
PCT APPLICATION NO.	PCT FILING DATE	U.S. APPLICATION NUMBERS ASSIGNED (if any)		

I hereby appoint the following attorneys and agent(s) to prosecute said application and to transact all business in the Patent and Trademark Office connected therewith and to file, prosecute and to transact all business in connection with international applications directed to said invention:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY (CONT'D)
(Includes Reference to Provisional and PCT International Applications)

Attorney's Docket No.

022701-892

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RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			
FULL NAME OF FIFTH JOINT INVENTOR, IF ANY		SIGNATURE	DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			
FULL NAME OF SIXTH JOINT INVENTOR, IF ANY		SIGNATURE	DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			
FULL NAME OF SEVENTH JOINT INVENTOR, IF ANY		SIGNATURE	DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			
FULL NAME OF EIGHTH JOINT INVENTOR, IF ANY		SIGNATURE	DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			